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The Effect of Energetic Charged Particle
Bombardment on the Properties of Thin Films
of High T_c Superconductors

Final Technical Report
ONT Postdoctoral Fellowship
10/1/88

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I. Introduction

The research that has come out of this past years ONT Postdoctoral Fellowship has varied greatly from what was planned in the original proposal due to the circumstances encountered. Basically, the research that has been completed began by measuring the radiation sensitivity (as determined by resistance versus temperature) of plasma-arc sprayed films, and later laser evaporated films, provided by sources outside NRL (specifically, SUNY-Stony Brook and AT&T Bell Laboratories, respectively). Next, measurements were performed on thin films produced at NRL by molecular beam epitaxy (MBE). Towards the end of this year the focus has been on actually making thin films by laser evaporation and also making radiation sensitivity measurements of the critical current. Specialized equipment has been constructed to make radiation damage measurements and also to make laser evaporated films. The basic design of this equipment is shown in Figs. 1 and 2. Also the two published papers describing radiation damage results have been attached.

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II. Summary of Current Understanding

A. Radiation Damage

Since the discovery of the new class of high T_c superconductors [1,2] much research has been published on the general properties of these materials including papers in the area of radiation effects [3-23]. To summarize the effects of radiation or the controlled introduction of disorder on thin films of these new materials it is required that we describe the superconducting pair wave function Ψ , as,

$$\Psi = \delta e^{i\phi}$$

where δ is the pair amplitude and ϕ is the phase. From this expression it is clear we can destroy the superconductivity by either reducing δ or destroying the phase coherence of ϕ .

For weakly coupled polycrystalline superconductors radiation damage evolves as the destruction of phase coherence between individual grains of good superconductor. Here the onset temperature of the superconductive resistive transition, $T_{c\text{-onset}}$, stays constant while the completion temperature, $T_{c\text{-completion}}$, is gradually lowered thus yielding a broad resistive transition. For better quality films, on the other hand, it is expected, based on the old superconductors [22], and recently seen [21], that δ is lowered through the controlled insertion of electron-defect scattering centers. Lowering δ evolves as a simultaneous lowering of $T_{c\text{-onset}}$ and $T_{c\text{-completion}}$.

Although both of the above regimes (δ reduction and loss of

ϕ coherence) of superconductivity destruction have been explored in the old high T_c superconductors by varying the morphology of deposited films [23,24], we do not have this luxury yet with the new materials due to the complexity involved in their formation and their short coherence length, ξ . Gross evidence supporting this comes from the order of occurrence of destruction regimes, i.e., loss of ϕ coherence followed by δ reduction. Due to the polycrystalline nature of ceramics in general, and the neighboring oxygen-deficient phases being either semiconducting or insulating rather than metallic, it is easy to see why prototype films would be weakly coupled and demonstrate loss of ϕ coherence on exposure to radiation. With improvements in film production films have recently been produced which show δ reduction indicating a better quality film [21]. Therefore, at least with the currently available films the term "quality" is still not one which is easily comparable, i.e., with the old high T_c compounds, the A-15's, the term "quality" could be used to correlate T_c as well as other physical properties with the residual resistivity [22].

B. Laser Evaporation Film Production

The fact that any effort towards film production has been made is a strong example of how far a research effort can separate from the initial proposal due to the circumstances encountered. In preparing this final report I discovered a letter to my advisor indicating my concern over the availability of films. Therefore, the problem of the availability of high

quality thin films was an expected problem from the start of this project.

At the 1988 March Meeting of the APS I heard many talks on the fabrication of high T_c thin films. The basic problem, as might be expected from the start, is controlled, epitaxial deposition of three different elements vaporized through various means, e.g., sputtering, e-beam evaporation, knudsen cells, etc. A simple solution to having to control three different sources is to only have one source and have your means of vaporization be indifferent to the three components. Laser evaporation or ablation offers such an alternative and was especially attractive due to the simplicity of design as shown in Fig. 2. The advantages and high quality results obtained from the laser evaporation deposition scheme were noted by myself at this conference.

In the laser ablation process a single pellet is used as the thin film source of material and the vaporization comes from a very intense and short burst of laser light. Since the early discovery of the advantages of this technique by Venkatesan's group at Bellcore [25] much work has been published in this area [25-32]. The laser evaporation system designed by myself and currently set up in the Laser Physics Branch at NRL is designed not only to allow numerous high quality thin films to be produced, but it also allows easy enhancement to deposition regimes not yet reported in the literature. From what is currently being published about thin film deposition it is an

important goal to be able to produce high quality superconducting-as-deposited films on silicon.

Currently, the results from the films which are being produced are very encouraging. These films are metallic in the normal region and show T_c -onset at 90K and zero resistance at $T \geq 50K$. The pellet used in this process has been adjusted several times to produce the desired 1-2-3 stoichiometry of the metallic elements. Also the pellet contains the fluoride of barium. This is as a precaution against forming barium carbonates at the grain boundaries which has been suggested as a possible cause of the weak link or weak coupling behavior often observed [32].

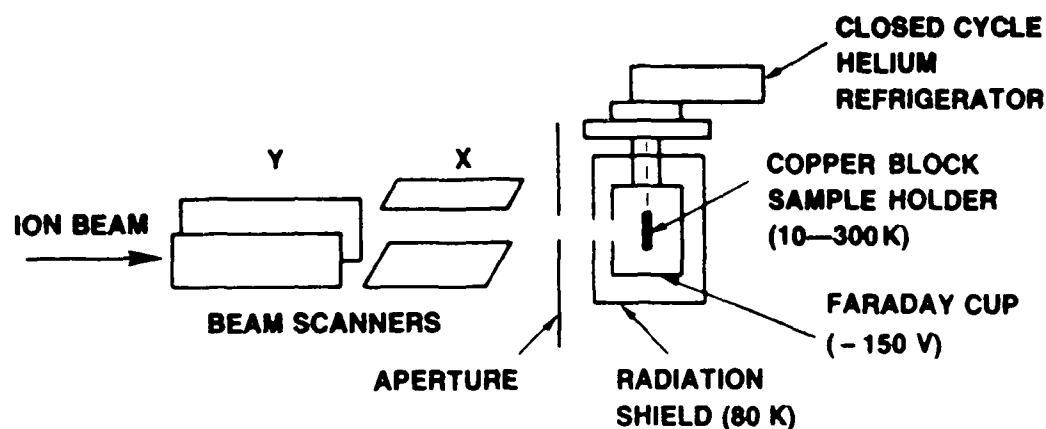
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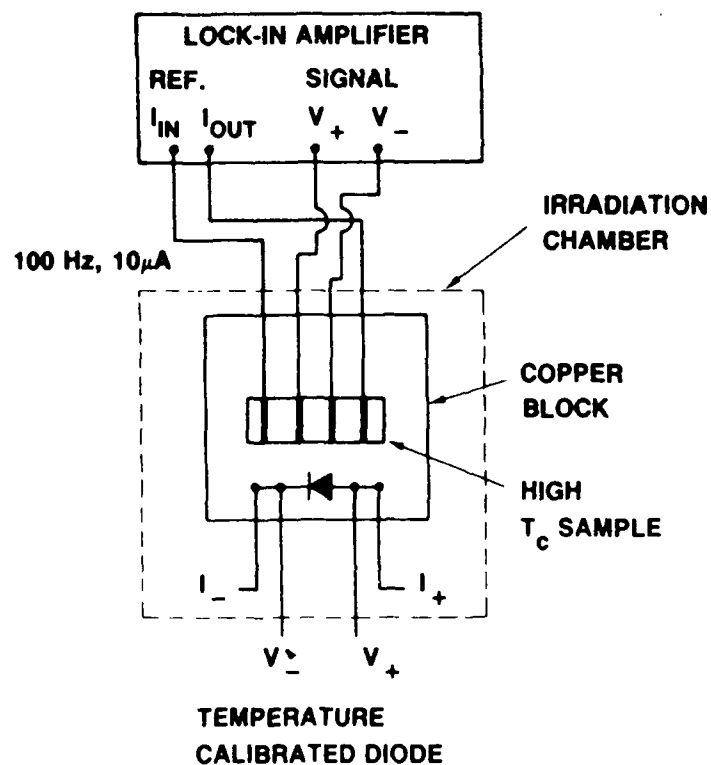
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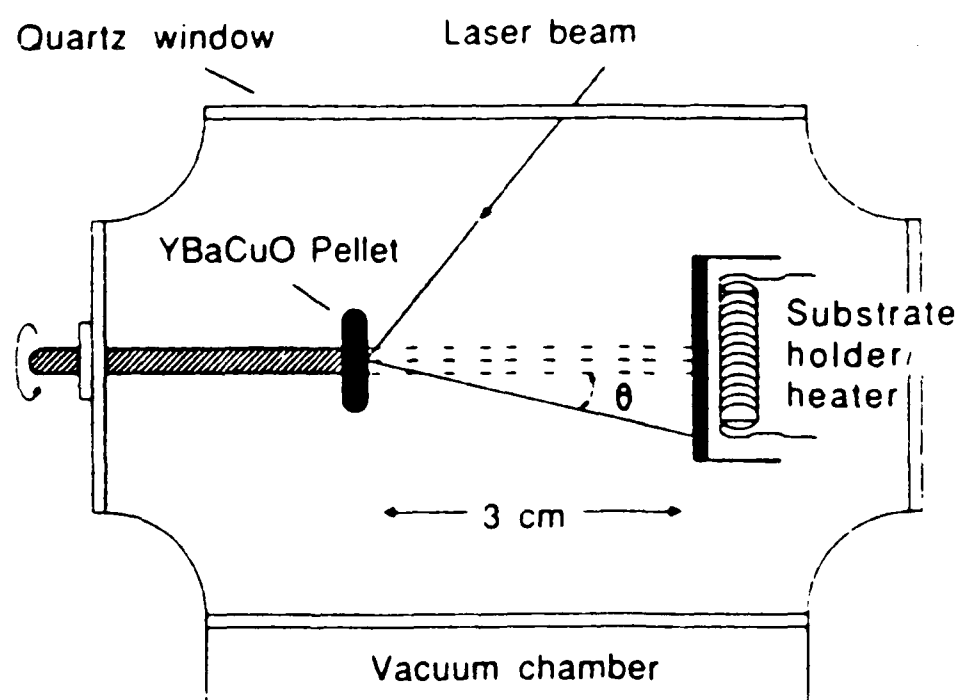
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IRRADIATION CHAMBER



RESISTANCE AND TEMPERATURE MEASUREMENT





Schematic of the deposition system.

Catastrophic loss of superconductivity in ion-irradiated films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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We have investigated the effects of low fluence ($< 10^{14} \text{ cm}^{-2}$) 63 MeV H^+ and 65 MeV He^{2+} irradiation of prototype thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ produced by a plasma-arc spray technique. The observed change in the resistance versus temperature behavior is much more dramatic than that observed for films produced by other techniques and resembles qualitatively a bond percolation threshold. The radiation sensitivity of these plasma-arc spray films is concluded to be due to poor intergranular characteristics. This information is being used to modify the processing steps to improve the properties of films produced by this technique. \rightarrow to p

The discovery of high T_c superconducting materials^{1,2} has prompted much research activity worldwide. Since successful applications in harmful radiation environments will require that the superconducting properties be maintained during irradiation, understanding the radiation sensitivity of these materials is an important consideration. This is especially the case since preliminary radiation damage measurements of e -beam (electron-beam) deposited³⁻⁵ and laser-evaporated⁶ thin films ($\sim 1 \mu\text{m}$), and sintered pellets⁷ of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ indicate a 1-2 order of magnitude increase in sensitivity as compared to the old high T_c compounds, i.e., the $A-15$ compounds. This increased sensitivity has been ascribed to the granular nature of the films and the presence of insulating behavior in other phases, and not to the intrinsic sensitivity of the bulk material.³⁻⁷ More recent measurements by our group on thick films ($\sim 200 \mu\text{m}$) of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ produced by a plasma-arc spray technique^{7,8} indicate a sensitivity much greater than that observed for the thinner e -beam and laser-evaporated films. Although the plasma-arc spray technique was in the early stages of development as applied to superconductors, tests of radiation sensitivity were performed to determine directions for improvements. In this letter we present new data showing this increased radiation damage sensitivity and account for it in terms of recent ideas about radiation-enhanced decoupling of the granules and a qualitative application of bond percolation theory.

The preparation of thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ by the plasma-arc spray technique is described in detail in Ref. 9. A large piece of plasma-arc spray film was cut on a low-speed diamond saw into seven samples of approximately the same

size and geometry ($2.3 \text{ mm} \times 7.4 \text{ mm}$). Three of these samples were irradiated with 63 MeV H^+ ions to fluences of 7.5×10^{12} , 2.06×10^{13} , and $7.5 \times 10^{13} \text{ cm}^{-2}$, respectively, and another three were irradiated with 65 MeV He^{2+} ions to fluences of 3.76×10^{11} , 1.03×10^{12} , and $3.75 \times 10^{12} \text{ cm}^{-2}$, respectively. Irradiations were performed at room temperature and in air at the UC-Davis cyclotron. After irradiation, silver paint electrical contacts were made and the resistance-temperature behavior was measured for each of the films. The data were then compiled to produce a single plot showing the progression of radiation damage for each type of irradiation, i.e., minor differences in resistance measurement geometry were ignored. The resistance measurement was made between room temperature and liquid-helium temperature using a four-point ac technique with a measurement current of $10 \mu\text{A}$.

The results for the H^+ - and He^{2+} -irradiated samples are shown in Figs. 1 and 2, respectively. Similar conclusions can be drawn from both sets of results. These are that (1) the T_c onset does not change with increasing particle fluence, (2) at low fluences T_c completion does not change significantly with increasing particle fluence with the exception of the most heavily irradiated samples, (3) the transition from the normal to the superconducting state occurs in one step, i.e., there is no sign of a second transition, (4) the room-temperature (RT) resistivity is very sensitive to radiation damage and increases linearly with increasing particle fluence, and (5) at approximately the same value of nonionizing energy deposition between the second and third irradiations, for both H^+ and He^{2+} , the zero-resistance state of the film as measured was lost catastrophically. All of the changes produced by the irradiations were due to displacement damage effects characteristic of ion bombardment because irradiation of a piece of the same material to a dose of

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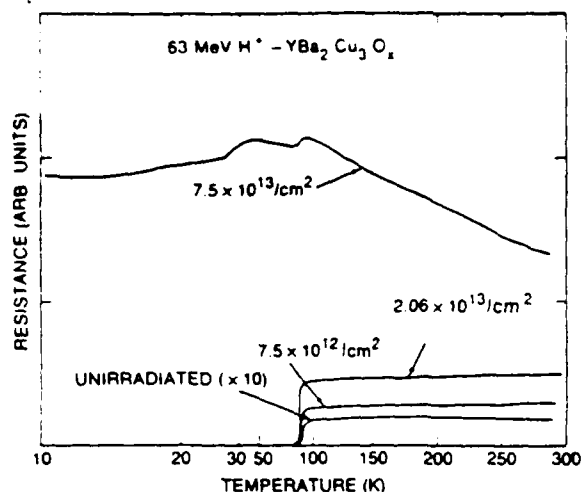


FIG. 1 Resistance vs temperature for $\text{YBa}_2\text{Cu}_3\text{O}_x$ plasma-arc sprayed films ($x \approx 7$) irradiated with 63 MeV H^+ ions. Note that the temperature axis is actually the temperature-sensing diode voltage which becomes extremely nonlinear below 40 K.

200 Mrad (Si) with 53 MeV e^- caused no appreciable change in either the RT resistivity or T_c [the largest 63 MeV H^+ and 65 MeV He^{2+} irradiations produced a dose of 14 and 8 Mrad (Si), respectively]. Furthermore, this increased radiation damage sensitivity is *not* seen for the laser-evaporated samples for similar fluences.⁶

The major differences in the radiation behavior between plasma-arc spray films and e -beam-deposited and laser-evaporated films are observations (3) and (4) above. The RT resistance increases faster than would be expected based on results on thinner films, and yet no second transition gradually lowering T_c completion with increasing fluence is seen. This means that an unperturbed superconducting path

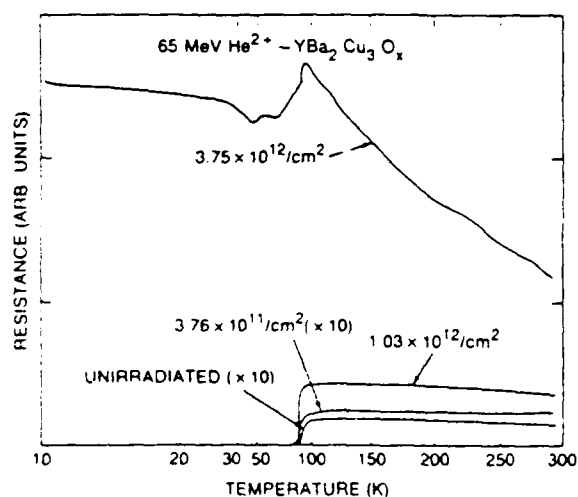


FIG. 2 Resistance vs temperature for $\text{YBa}_2\text{Cu}_3\text{O}_x$ plasma-arc sprayed films ($x \approx 7$) irradiated with 65 MeV He^{2+} ions. Note that the temperature axis is actually the temperature-sensing diode voltage which becomes extremely nonlinear below 40 K.

continues to exist through the sample even after the first two irradiations. This path shorts out the rest of the sample below T_c as the RT resistivity is rapidly increasing with fluence above T_c .

We attribute the difference in the radiation behavior between the plasma-arc spray films and the e -beam-deposited and laser-evaporated films to the different morphology of the films. The plasma-arc spray films studied were relatively open, cracked, and irregular structures less than 80% dense and consisting of randomly oriented granules approximately 10 μm across. e -beam-deposited and laser-evaporated films consist of more uniform and closer packed granules about 1 μm in diameter and have the c axis preferentially oriented perpendicular to the substrate and thus to the sample resistance measurement geometry. The critical current J_c , a property which is strongly dependent on the intergranular characteristics of the films, is evidence supporting the microscopic differences in film morphology. e -beam-deposited and laser-evaporated thin films have values of J_c on the order of 10^6 A/cm^2 , whereas these plasma-arc spray films have values of J_c less than 10^5 A/cm^2 .

The existence of an unperturbed, continuous superconducting path through the sample, which is present until it is destroyed by a small incremental particle fluence (or energy deposition), suggests that it would be instructive to view the damage results in the light of bond percolation theory. In particular, the observed catastrophic loss of superconductivity is strongly reminiscent of a percolation threshold. Treating the superconducting path between the voltage electrodes as forming an infinite cluster, a bond percolation system can be defined. Broken bonds between the clusters are formed by nonsuperconducting junctions between individual grains due to an insulating amorphous layer, structural defects, lowered oxygen content at the grain boundaries, or the presence of a different phase. Particle irradiation increases the number of nonsuperconducting or broken bonds until the superconducting path is finally destroyed by a small incremental fluence. At this point the zero resistance of the sample would be lost catastrophically, as is observed. However, a drop in resistance with decreasing temperature would still occur at T_c onset because most of the final unperturbed superconducting path would still be intact. Indeed the magnitude of the resistance drop might be expected to be comparable to that observed just prior to the incremental fluence that finally breaks the superconducting path. This effect can be seen in Fig. 2 and to a lesser extent in Fig. 1. Data suggesting that the volume fraction of superconducting material remains unchanged, even though radiation destroys a zero-resistance state, have also been seen elsewhere.^{4,6,7}

The similarity between the sudden loss of superconductivity observed in these films and other percolation phenomena is very compelling.¹⁰⁻¹¹ What is particularly interesting is that the percolation threshold appears to be reached by the deposition of a certain nonionizing energy, independent of the incident particle type. Assuming there is a percolation threshold, calculated values of the energy loss can be used to narrow the range of critical fluences. The calculated nonionizing energy loss for 63 MeV H^+ is 3.40 keV cm^2/g (see Ref. 14). Current estimates of the ratio of nonionizing energy loss

by 65 MeV He^{2+} ions relative to 63 MeV H^+ ions is 5.4–5.7, and from Figs. 1 and 2 the measured minimum critical fluence ratio was $2.06 \times 10^{11} / 3.75 \times 10^{11} = 5.5$. Using 5.5 from above as a minimum and 5.7 from the energy loss as a maximum, the range of critical fluences required to destroy superconductivity for incident 63 MeV H^+ and 65 MeV He^{2+} can be narrowed to $2.06\text{--}2.14 \times 10^{11} \text{ cm}^{-2}$ and $3.61\text{--}3.75 \times 10^{11} \text{ cm}^{-2}$, respectively. This means that the threshold was reached just after the second H^+ irradiation, whereas it was reached just before the end of the third He^{2+} irradiation. The degradation of the resistive drop of the largest fluence H^+ irradiation at T_c onset with respect to that for the He^{2+} irradiation is evidence supporting this idea.

In heavier ion irradiation of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films Clark *et al.*³ demonstrated a higher sensitivity based on the mean amount of nonionizing energy deposited per atom when the zero-resistance state was lost. Clark *et al.*'s value of approximately 1 eV/atom has since been obtained elsewhere with a wide range of ions and energies.^{4,6} On the other hand, the amount of energy deposited for the largest fluence on the plasma-arc spray films is on the order of 10^{-6} eV/atom!

We have chosen to apply the bond percolation theory to explain our results because of its success in describing transport phenomena in a wide range of situations such as composite media,¹⁰ amorphous solids,¹¹ and granular superconductors.¹² Furthermore, the percolation-like threshold in the effect of radiation on the transport properties of the plasma-arc spray films reemphasizes the importance of obtaining good electrical conductivity in the intergranular regions. With this in mind, the plasma-arc spray technique is being

improved to produce denser and mechanically stronger films with full oxygenation at the grain boundaries.

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THE INFLUENCE OF RADIATION DAMAGE ON THE SUPERCONDUCTING
PROPERTIES OF THIN FILM $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ⁺

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A.R. Knudson¹, and E.A. Burke²

ABSTRACT

We report measurements of the effects of high energy (~60 MeV) H and He ion irradiation of thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ produced by plasma-arc spray and laser evaporation, and of low energy (~10 MeV) B and N ion irradiation of the laser evaporated films. The observed changes in the resistance versus temperature behavior for H and He ion irradiation of the plasma-arc spray films are much more dramatic than that observed for films produced by other techniques and resembles qualitatively a bond percolation threshold. In contrast, high energy H and He ion irradiation of the laser evaporated films resulted in a negligible change in the resistance-temperature behavior. The low energy B and N ion irradiation of the laser evaporated film had the effect of a gradual lowering of the completion temperature of the superconducting transition (T_c -completion). The gradual lowering of T_c -completion with fluence, $d(\Delta T_c)/d\phi$, was found to scale with the nuclear energy loss and agreed well with similar results by other groups. This behavior is interpreted as a progressive decoupling of the grains and extrinsic to that of the bulk $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. *phi*

delta T sub c

51 C

(7-delta)

⁺Supported, in part, by the Naval Research Laboratory High Temperature Superconductor Program.

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²Mission Research Corporation, San Diego, CA

Introduction

Many of the applications of the recently discovered high temperature superconductors^{1,2} such as $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, especially uses in microelectronics, will involve the use of thin films. The radiation hardness of devices made from these materials will be determined largely by the radiation response of the films themselves. Therefore, it is important to determine the radiation damage processes involved and the factors that limit the hardness of these new materials.

It was recently shown by our group that although both high energy electrons and protons increased the normal resistance of a thin ($\sim 150\mu\text{m}$) film of the high temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, neither particle affected the onset temperature of the superconducting transition, T_c -onset, up to the fluences used, which corresponded to typical space mission exposures³. The films used in this earlier study were made by the plasma-arc spray technique. Because of the difficulty involved in controlling the parameters of the fabrication process, the radiation sensitivity of plasma-arc spray films and films made by other techniques, such as electron beam (e-beam) or laser evaporation varies strongly from sample to sample^{3,4,20}. However, the early films made by the plasma-arc spray technique were found to be many orders of magnitude more radiation sensitive than thinner films made by e-beam or laser evaporation⁴. In fact, it appears that to some extent the radiation sensitivity of a film is a measure of the overall quality, e.g., porosity, cracking, full oxygenation at the grain boundaries, etc.

In this paper we describe the inherent difference in radiation sensitivity of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films produced by plasma-arc spray and laser evaporation. It is observed that for the plasma-arc spray films studied, the deposition of a relatively small level of nonionizing energy ($\sim 10^{-5}\text{eV/atom}$) resulted in the catastrophic loss of the zero resistance state. Nonionizing energy deposition is defined as the total amount of energy deposited by the primary particle and each generation of recoils which results in kinetic energy of the nuclei of the target. The progression of radiation damage with fluence to the point of the catastrophic loss of superconductivity has the qualitative appearance of a bond percolation threshold. For the laser evaporated films, on the other hand, the damage evolves as a progressive decoupling of grains similar to that found for e-beam films by other groups⁵⁻⁷.

New calculations of the nonionizing energy deposited by high energy H and He ions will be discussed in the New Calculations section. These calculations include elastic and inelastic interactions, and the Lindhard energy partition. In addition, the implications of the results presented for the application of high temperature superconductors in radiation environments, including space, will be discussed.

Experimental Details

The series of sample irradiations to be described were carried out at two different accelerators. High energy (~ 60 MeV) H and He ion irradiations were done at room temperature in air at the Crocker Nuclear Laboratory cyclotron at UC-Davis and the lower energy (~ 10 MeV) B and N ion irradiations were done at room temperature in a specially designed radiation chamber at the Naval Research Laboratory (NRL) 3 MeV Tandem Van de Graaf. At UC-Davis the particle current was monitored by collecting the secondary electrons emitted from an aluminum foil. This monitoring system was calibrated before each run with a Faraday cup current measurement. The calibration was repeatable to $\sim 2\%$ and accurate to $\sim 10\%$. A thermoluminescent detector array was used to determine the beam profile. The fluences used in these irradiations were less than 10^{14} particles/cm². The fluences used in the low energy NRL irradiations were less than 3×10^{15} particles/cm².

Measurements of the change in the transport properties with nonionizing energy deposition have been centered on studying the resistance as a function of temperature, $R(T)$. This measurement is carried out using an A.C., four point probe technique at 100 Hz with a $10 \mu\text{A}$ sampling current. The temperature was measured by attaching the superconducting sample in good thermal contact with a temperature-calibrated diode. The temperature of the sample and diode was varied either by slowly lowering them into a dewar of liquid helium or by attaching both to a closed-cycle helium refrigerator. The resistance as a function of temperature was recorded on an x-y recorder. The change in the diode resistance with temperature is linear for temperatures from room temperature (300 K) to about 30 K, but below 30 K the resistance becomes nonlinear with temperature as demonstrated in Figs. 1-4.

Thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ prepared by the plasma-arc spray technique and the laser evaporation technique are described in detail in ref. 9 and 10, respectively. The thicknesses of the plasma-arc spray films were $\sim 200 \mu\text{m}$, whereas the laser evaporated films were on the order of $1 \mu\text{m}$ thick. For logistical reasons the experiments done using the UC-Davis cyclotron and the NRL Van de Graaf were different. For the UC-Davis experiments the original piece of superconducting material was cut into four pieces of roughly equal size ($\sim 2 \times 8 \text{ mm}^2$). Each piece was irradiated at UC-Davis to a different total fluence with one piece receiving no irradiation. The films were brought back to NRL and $R(T)$ was measured for each fluence (piece) and compiled to produce a single plot showing the progression of radiation damage, i.e., small differences in sample size and measurement geometry were ignored. For low energy B and N irradiations performed at NRL, $R(T)$ was measured as a function of dose on a single sample.

The general morphology of the two sets of films was extremely different. The plasma-arc spray films studied were open porous films having extensive cracking, and a mean grain size on the order of $10 \mu\text{m}$. The laser evaporated films studied, on the other hand, had smaller grains about $1 \mu\text{m}$ in size and

contained no cracks.

Results

Three series of irradiations were carried out. The first two were high energy H and He ion irradiations of plasma-arc spray and laser evaporated films. The results for the plasma-arc spray films are shown in Figs. 1 and 2. For high energy H and He ion irradiations of laser evaporated films, up to a total fluence of $3 \times 10^{13}/\text{cm}^2$ for both ions, there was no observable change in the $R(T)$ characteristics. For the plasma-arc spray film irradiations the fluence intervals were chosen so that the energy deposition, based on Rutherford scattering alone, would be similar for H and He ions. As is evident from the similarity of Figs. 1 and 2, the radiation damage is correlated with the amount of energy deposited into displacement damage. Furthermore, the amount of nonionizing energy deposition required to destroy the superconducting zero resistance state in the plasma-arc spray films is very small.

The important features common to both Fig. 1 and 2 are that: (1) T_c -onset does not change with increasing particle fluence, (2) at low fluences T_c -completion does not change significantly with increasing particle fluence, with the exception of the most heavily irradiated samples, (3) the transition from the normal to the superconducting state occurs in one step, i.e., there is no sign of a second transition, (4) the room temperature resistivity is very sensitive to radiation damage and increases approximately linearly with increasing particle fluence, and (5) at approximately the same value of nonionizing energy deposition between the second and third irradiations for both H and He ion irradiation, the zero resistance state was lost catastrophically⁴. The requirement of a certain level of energy deposition or defect density to destroy the zero resistance state resembles a threshold.

Since the laser evaporated films were irradiated to similar or greater fluences of either high energy H or He ions compared to the plasma-arc spray films and only a negligible change in the $R(T)$ characteristics was observed, the difference in radiation sensitivity for the two types of films must be inherent to the method of preparation. In order to see a change in the $R(T)$ transport properties of the laser evaporated films the level of nonionizing damage was increased through the use of heavier low energy ions. The results for 6.41 MeV B^{2+} and 9 MeV N^{2+} are shown in Figs. 3 and 4, respectively. In this case, the fluences were chosen to be the same so that the difference in radiation response would be a reflection of the difference in nonionizing energy loss for the two ions. The level of change in transport properties is consistent with a mechanism based on the relative level of displacement damage.

The important features common to both Figs 3 and 4 are that: (1) T_c -onset does not change with increasing particle fluence, (2) at low fluences T_c -completion (the temperature at which the film reaches a zero resistance state) changes with increasing

particle fluence, and 3) the room temperature resistivity increases approximately linearly with particle fluence, but at a much slower rate than that found for the plasma-arc sprayed films. It should be noted that low energy irradiations, such as those in Figs. 3 and 4, could not be done for the plasma-arc spray films due to the small range of the particles.

New Calculations

The approach used to calculate nonionizing energy loss (NIEL) by 63 MeV H and 65 MeV He ion in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ is an updated version of that previously applied to proton bombarded Si and GaAs^{22,23}. The calculations include all the interactions that contribute to NIEL. These are: Rutherford (Coulombic), nuclear elastic and non-elastic cascade evaporation processes. The partition of primary recoil energy into ionizing and nonionizing components was accounted for by applying the Lindhard model²⁴. Details concerning all aspects of the approach used in the present method of calculation are provided in ref. 25.

As described in ref. 25, at a given recoil energy, the high atomic number elements lose a greater fraction of their energy in nonionizing processes. In view of the elements in the superconducting compound in the present study, the computed results for NIEL are expected to be sensitive to details of the calculation. The greatest uncertainty is associated with the cascade-evaporation interaction which produces recoils with energies that are typically 1000 times greater than is the case for the Coulombic interactions.

At the energies considered here the calculated contribution from the inelastic interactions to NIEL was found to be almost twice as large as that for the elastic interactions. The value of NIEL found for 63 MeV H was $3.40 \text{ keV-cm}^2/\text{g}$ and that for 65 MeV He ions was 5.7 times greater.

Discussion

Since the relatively recent discovery of a new class of high temperature superconducting materials^{1,2}, a large amount of research has been published including papers in the field of radiation effects^{3-7,11-20,26,29}. The results presented for the plasma-arc spray films are unique due to their heightened sensitivity. The progression of radiation damage in these films, demonstrating that a certain level of energy deposition is required to destroy the zero resistance state, qualitatively resembles a bond percolation threshold. In this application a bond percolation threshold can be thought of as a long single thread of good superconductor existing between the voltage probes until a certain energy deposition (the bond percolation threshold) is reached at which point the thread is broken and a nonzero resistance will result.

The gradual change in T_c -completion ($T_c(R=0, \phi=0) - T_c(R=0, \phi>0)$ or ΔT_c -completion) with increasing particle fluence, similar to that in Figs. 4 and 5, was first noted by White et al.⁷ as being

indicative of the progressive decoupling of superconducting grains. For a granular superconducting film, like these used here, the coupling between individual grains occurs via the Josephson interaction once the Josephson coupling energy, E_J , is greater than the thermal energy, $k_B T$, where k_B is Boltzmann's constant²¹. Since the Josephson coupling energy is inversely proportional to the normal state resistance, R_N , of the junction between two grains, it is possible to understand in a purely qualitative way, how displacement damage, which increases R_N , would lead to a decrease in T_C -completion. An increase in R_N would decrease the coupling energy, E_J , and thus lower the temperature to which the grains must reach in order to satisfy the requirement $E_J > k_B T$, where $T = T_C$ -completion and allow a zero resistance state to occur between the grains. White et al.⁷ noted a change in T_C -completion with fluence for MeV ions covering nearly two orders of magnitude in nonionizing (or nuclear) energy loss. Furthermore, it was suggested that $d(\Delta T_C)/d\phi$ was a measure of the damage, and that this quantity scaled with the nuclear energy loss. In Fig. 5 we plot $d(\Delta T_C)/d\phi$ for the laser evaporated films of Figs. 4 and 5, and compare it to similar data by White et al.⁷ and Clark et al.⁵. The straight line drawn is based on White et al. data since it is the most complete set of data currently published. The close agreement between the damage factor, $d(\Delta T_C)/d\phi$, for our laser evaporated films with those found by White et al.⁷ and Clark et al.⁵ demonstrates that a similar mechanism is involved, i.e., a progressive decoupling of superconducting grains. Fig. 5 should not be viewed as a universal curve for relating the damage factor $d(\Delta T_C)/d\phi$ with nonionizing energy loss for all films. Although granular films made by other techniques would be expected to have the same linear dependence, the actual values of damage factors for a given nonionizing energy loss would be different²⁰.

The radiation sensitivity of granular $YBa_2Cu_3O_{7.6}$ films is, in its early stages, a result of the intergranular characteristics rather than that of the bulk material³⁻⁷. The crystalline to amorphous transition takes place long after the film has lost its low temperature zero resistance state^{6,7}. Although the details of the mechanism whereby radiation damage affects the grain boundaries is not yet known, it was suggested that incident particles cause displacement damage in the material and that defect migration is possibly involved⁶. Preliminary^{7,16,29} low temperature measurements indicate a slightly higher radiation sensitivity at low temperatures with noticeable recovery near 200K on warming¹⁶ thus eliminating defect migration to the grain boundaries as a possible contributor. Measurements of radiation damage as a function of temperature are planned for the future to better understand the role of defect migration and room temperature annealing. A film with poor intergranular structure, such as cracking, low density, or incomplete oxygenation at the grain boundaries, will be more sensitive to radiation than a high density film with good characteristics. Scanning electron microscopy of the plasma-arc

spray films show that this is indeed the case. Further evidence supporting the poor intergranular characteristics of the plasma-arc spray films comes from the critical current, J_c , of these films. The critical current is a quantity which is extremely sensitive to the intergranular properties and is three to four orders of magnitude lower for the plasma-arc spray films as compared to the laser evaporated films.

The implications of these results for space applications are quite clear. For applications requiring that thin films be functional in potentially harmful radiation environments, the thin films currently produced by e-beam evaporation and laser evaporation are suitable and will likely survive typical expected exposures. The plasma-arc sprayed films currently available have improved properties compared to the films used in this study and thus the possibility of their use in radiation environments should not be ruled out.

Conclusions

The measurements presented here are unique in that we have compared the radiation sensitivity of films produced by two different techniques and compared the sensitivity (for the laser evaporated films) with similar results by other groups using $d(\Delta T_c)/d\phi$ as a measure of the radiation damage. The results demonstrate that the radiation sensitivity of currently available films is an intergranular phenomena extrinsic to that of bulk $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and a strong function of processing technique. Therefore, as the quality of the intergranular region improves in polycrystalline films available for radiation damage measurements, perhaps through the addition of noble metals^{27,28}, or through the fabrication of single crystals, the radiation sensitivity should approach that of bulk $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. And if single crystal material is available the damage will evolve with fluence, at least in its early stages, as a reduction in T_c -onset similar to what was observed³⁰ and predicted³¹ for the A-15's. It is still interesting, though, that the plasma-arc spray films available when these experiments were done, could have such a radical difference in radiation sensitivity as compared to films made by other techniques such as laser or e-beam evaporation.

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FIGURE CAPTIONS

Figure 1

Resistance versus temperature for $\text{YBa}_2\text{Cu}_3\text{O}_x$ plasma-arc sprayed films ($x \approx 7$) irradiated with 63 MeV H^+ ions.

Figure 2

Resistance versus temperature for $\text{YBa}_2\text{Cu}_3\text{O}_x$ plasma-arc sprayed films ($x \approx 7$) irradiated with 65 MeV He^{2+} ions.

Figure 3

Resistance versus temperature for $\text{YBa}_2\text{Cu}_3\text{O}_{7.6}$ laser evaporated film irradiated with 6.41 MeV B^{2+} ions.

Figure 4

Resistance versus temperature for $\text{YBa}_2\text{Cu}_3\text{O}_{7.6}$ laser evaporated film irradiated with 9 MeV N^{2+} ions.

Figure 5

The change in T_c -completion with fluence ($d(\Delta T_c)/d\phi$) versus the nuclear or collisional energy loss, $(dE/dx)_n$, for the laser evaporated films studied here as well as results from other groups on e-beam films^{5,7}.

